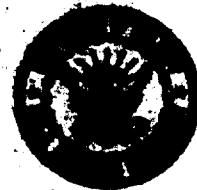


Modification of the Air/Sea Interface by Artificial Sea Slicks

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ABSTRACT

The physical properties of the air/sea interface may be significantly modified by adsorbed monomolecular films of surface-active materials. Damping of capillary waves, retardation of evaporation, inhibition of microconvectional cells, and modification of air-bubble properties are some of the microscale surface parameters affected by these monomolecular films. The concentration of airborne salt condensation nuclei produced from bubbled seawater is also altered by surface-chemical effects. These microscale effects can influence larger scale geophysical parameters, such as the transmission and reflection of light from the sea surface, the production of condensation nuclei by bursting bubbles, and the diminution of waves.

While natural sea slicks may occasionally exist and alter sea-surface properties, artificial surface films, which are more durable and effective in modifying air-sea interactions, have been selected on the basis of a series of field and laboratory experiments. Optimum ocean performance was demonstrated by oleyl alcohol, a chemically unsaturated fatty alcohol which forms a rapidly spreading, coherent monomolecular film, produces large capillary-wave-damping effects, and does not interact chemically with seawater.

PROBLEM STATUS

This is an interim report; work on this problem is continuing.

AUTHORIZATION

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MODIFICATION OF THE AIR/SEA INTERFACE BY ARTIFICIAL SEA SLICKS

INTRODUCTION

The interaction between the atmosphere and bodies of water can be significantly altered with a small quantity of material through the formation of an artificial sea slick. These sea-surface films are monomolecular layers of water-insoluble organic compounds. At present, the primary use of these one-molecule-thick films in the environmental control field is to retard evaporation of water from reservoirs (1,2). Only molecules with a special geometry can reduce the transport of water vapor across the air/water interface. For example, hexadecyl alcohol, one of the most effective water-insoluble compounds, has a linear molecular structure, and its molecules are able to pack tightly into a vertical orientation when the fatty alcohol monolayer is under high lateral compression (film pressure). Since this tightly packed structure is essential for evaporation retardation, many monolayer-forming substances that do not adenilate are not effective and can substantially destroy the efficiency of an alcohol film when they are present as impurities in small concentrations (1). Bending or branching of the normally linear hydrocarbon portion of the molecule or ionization of the hydrophilic group prevents close packing and drastically decreases the resistance to water-vapor transport (3).

It is unlikely that any monolayer spread onto the sea surface would significantly reduce the rate of evaporation. The wave-induced dilations and compressions of the sea surface would cause collapse and expansion of the surface film, thereby destroying its properties as a physical barrier to evaporation. Furthermore, the introduction of the natural surface-active organic material of the sea into the artificial film would decrease its efficiency as a vapor barrier due to the surface adsorption of nonlinear unsaturated fatty compounds common to the ocean (4).

However, the spreading of monomolecular films on the surface of the sea can significantly affect several other properties of the air/sea interface. For example, alteration of the sea-surface temperature, the inhibition of microconvectional cells (5), and the damping of eddies at the free surface of a turbulent liquid (6) are caused by monomolecular layers of organic compounds. In addition, significant capillary-wave-damping effects have been demonstrated both in the laboratory and at sea as a result of naturally occurring films (sea slicks) (7). Natural oceanic surface-active materials also modify the stability (8) and bursting properties (9) of air bubbles at the sea surface and alter the concentrations of salt condensation nuclei produced from bubbled seawater (10). These effects become most pronounced when the molecules of the film are compressed to a point where the monolayer possesses the properties of a relatively incompressible, two-dimensional liquid or solid. The surface layer is thereby immobilized, and the fluid dynamics of the air/sea interface is substantially altered.

The ultimate goal of this problem is the useful alteration of the ocean and its environment by an artificial sea slick spread onto the sea in a predetermined location. Once applied to the sea surface, the film must be able to withstand wind and wave activity (2), biological feeding (11), and photochemical oxidation by ultraviolet radiation (12). Furthermore, the film-forming material must be compatible with the biological systems of the sea (13) and not contribute to marine pollution.

Several chemical criteria must be considered in the selection of a monolayer-forming material for the creation of a durable, effective artificial sea slick. The molecule must

be polar and nonpolar in structure, containing a hydrophilic group to anchor it to the water surface and a hydrophobic hydrocarbon chain. The material must spread spontaneously into a water-insoluble monomolecular film so that only small quantities of material are needed to affect large surface areas. The hydrocarbon portion of the molecule should contain at least 18 carbon atoms to reduce evaporation and dissolution of the one-molecule-thick layer. Commercially available materials which satisfy these requirements are fatty acids, alcohols, and esters (vegetable oils) derived from natural products. A large number of synthetic surface-active agents are also available at greater cost, but they may raise questions with regard to water pollution.

Preliminary experiments were performed in the laboratory and on the Chesapeake Bay to determine (a) the feasibility of creating and sustaining a monomolecular film on the sea, (b) the resistance of various monolayer-forming materials to dispersive forces, (c) the quantities of surface-active material required, and (d) the most effective monolayer-forming chemicals for the creation of a durable artificial sea slick of potential usefulness.

EXPERIMENTAL FIELD OBSERVATIONS

The goal of the first field experiment was the generation of an artificial slick of controlled dimensions with a known quantity of material. A highly visible surface film of oleic acid was formed on the Chesapeake Bay, 400 m east of Ravelle Cliffs, Maryland. While spreading the surface-active material, it was found that a stripe of artificial slick could be left in the wake of the motor launch which clearly marked the route traversed (Fig. 1). A large rectangular slick was produced by laying a series of parallel stripes 800 m long and 20 m apart. Four liters of oleic acid created a surface film which measured 480 by 800 m (Fig. 2).

The oleic acid was spread at a rate of 75 ml/min from a pressurized container while the motor launch moved at a constant speed of 5 knots. New stripes of slick were laid on the windward side of the main body of material perpendicular to a 2.7-m/sec wind. The intrinsic spreading rate of the monolayer, in addition to the motion of the surface water caused the stripes to merge with the main slick in about 15 min. The total time of application was 1 hr and 46 min. The slick was more expanded in the shallower water near shore than at its eastern end where it was confined by a current flowing southward in opposition to a southerly wind. The slick remained coherent and drifted slowly north, its shape and speed being controlled by surface-water drift and wind.

About 6 times as much material was required to form the slick than the amount calculated for a monomolecular layer of similar size. This material inefficiency can be accounted for by two factors. Whenever excess oil was applied to the water surface, it did not immediately spread but existed as a bulk lens of liquid in equilibrium with its monolayer. This excess oleic acid represented a reservoir of monolayer-forming material to cover new surface areas or to repair breaks in the slick caused by surface dilations and losses from the monolayer. A second explanation for the monolayer inefficiency was the loss of the fatty acid from the surface through its interaction with the slightly alkaline Bay water (pH = 8.0). The oleic acid was saponified to water-soluble soaps which were easily expelled from the surface by a number of processes.

Another significant observation was obtained from a simultaneous comparison of a cottonseed oil slick (mixed glyceryl esters) with one made from equal quantities of oleic acid. The cottonseed oil film was estimated to cover an area 4 times greater than that of the oleic acid 2 hr after spreading, since a sizable portion of the fatty acid film had been lost by interaction with the seawater. However, damping in the cottonseed oil film was less intense than in the oleic acid monolayer. More wave structure was visible in the cottonseed oil slick, and its reflectance anomaly was less pronounced, having the appearance of a natural slick (film pressure 15 dynes/cm or less).



Fig. 1 - Artificial oleic acid slick generated in specific pattern
on the Chesapeake Bay



Fig. 2 - Artificial slick (0.38 km^2) generated from oleic acid
on the Chesapeake Bay

In laboratory experiments, these two substances damped capillary waves to the same degree when their monolayers were at low film pressures. However, in the field these films were at their maximum spreading pressure in equilibrium with bulk, unspread droplets of material. The film pressure of the fatty acid slick (30 dynes/cm) was twice that of the cottonseed oil film (15 dynes/cm). It is likely, therefore, that the resistance of a slick to wave formation is not entirely related to its wave-damping characteristics but is also a function of its film pressure (surface tension decrease), which governs the wavelength produced by a particular wind generated disturbance.

Adsorbed surface-active material not only attenuates existing waves but also inhibits wave formation (14,15). According to Miles (16), the addition of a sufficient quantity of a soluble surface-active agent to produce an inextensible film at the air/sea interface can increase the minimum wind velocity necessary to produce waves by almost one order of magnitude. While natural sea slicks should in theory withstand winds of this order (12 m/sec), their constituents are dispersed at sustained wind speeds of 7 to 9 m/sec. The concentration of active material in the underlying seawater is insufficient to supply and maintain a coherent film under highly dispersive conditions.

However, the artificial films of water-insoluble compounds may possess the wave-inhibiting properties against winds as high as 12 m/sec, as postulated by Miles (16). On several occasions artificial slicks were formed on the Chesapeake Bay when winds were 7 to 10 m/sec. The Bay was quite choppy with 1-m swells and white caps, and all natural slicks had been dispersed. Even under these adverse conditions, coherent artificial surface films could be maintained as long as excess surface-active material was present to repair incipient ruptures in the slick. Films formed from pharmaceutical-grade oleyl alcohol at this sea state possessed the rapid spreading and high capillary-wave-damping properties of oleic acid and the durability of cottonseed oil films.

FILM STABILITY ON SEAWATER

Since loss of a monolayer can occur from saponification and dissolution, laboratory experiments were performed to compare the durability of several materials in monomolecular form on slightly alkaline substrates. Monolayers of oleic acid (white, distilled, Mann Research Laboratories), oleyl alcohol (white, Hormel Institute), cottonseed oil (USP), and peanut oil (commercial) were spread on three different substrate solutions: distilled water, seawater (pH 8.4), and sodium tetraborate solution (2.5 g/l $\text{Na}_2\text{B}_4\text{O}_7 \cdot 5\text{H}_2\text{O}$, pH 9.1). The film pressures were measured with a Cenco hydrophil balance mounted on a paraffin-covered glass tray 70 by 12 cm and 1 mm deep. The monomolecular films were compressed to selected film pressures with a glass barrier coated with paraffin wax.

The film pressure was maintained at 10.3 dynes/cm for cottonseed oil and peanut oil and at 20.7 dynes/cm for oleic acid and oleyl alcohol. These film pressures are in the steep portion of the respective film-pressures-vs-area curves (Fig. 3). Since film pressures in this region change substantially with small changes in the number of molecules of film per unit area of surface, monolayer losses were readily detected. A modified syringe pump was used to drive the waxed barrier and maintain a constant film pressure. The barrier could be driven at various speeds ranging from 0.005 mm/min to 77.4 mm/min, thereby providing extremely small decreases in surface area to compensate for film losses. The rate of loss of material from the surface was measured as the decrease in film area with time at constant film pressures.

The substrate solution was aged in the hydrophil tray for at least half an hour. Its surface was then swept three times with the paraffin-covered bar to remove surface-active contaminants. A small quantity (0.05 mg) of the monolayer-forming material contained in a spreading solvent was added to the surface with a microsyringe, and the barrier was driven forward rapidly until the selected film pressure was reached. The

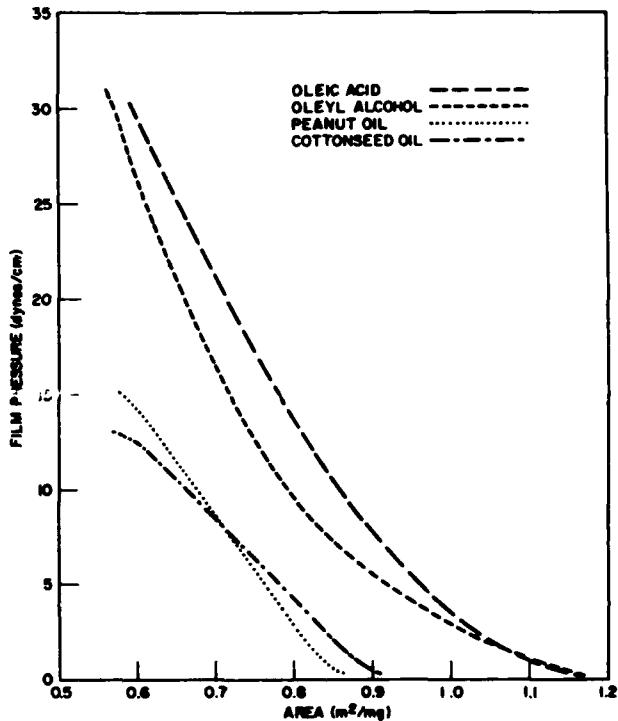


Fig. 3 - Dependence of film pressure on area occupied by monolayers of various pure compounds and vegetable oils on distilled water

timer was then started, and the barrier was advanced very slowly to maintain a constant film pressure. After an initial 5-min period, the time and the corresponding position of the barrier was recorded. The first reading was taken as the 100% value. From the change in position of the barrier, the percent of spread film remaining at the surface at any particular time could be calculated.

The film pressures of the oleyl alcohol and cottonseed oil monolayers remained constant for at least 30 min, and there was no detectable loss of surface-active material. Both the oleic acid and the peanut oil monolayer showed loss of material from the surface with time, as indicated by graphs of the percent of material remaining at the surface vs monolayer age (Figs. 4 and 5). Material lost from the peanut oil monolayer (Fig. 5) was probably due to saponification of the free fatty acids which are constituents of the vegetable oil. Since the rate of loss of material from the oleic acid and peanut oil films increased with increasing pH, the major cause of film loss from an artificial sea slick made with these materials would appear to be saponification and dissolution of the materials by the alkaline seawater. Thus, fatty acids are not the best substances from which to create an artificial slick of long duration, whereas a fatty alcohol or pure fatty ester (glyceryl triester) does not interact significantly with alkaline substrates.

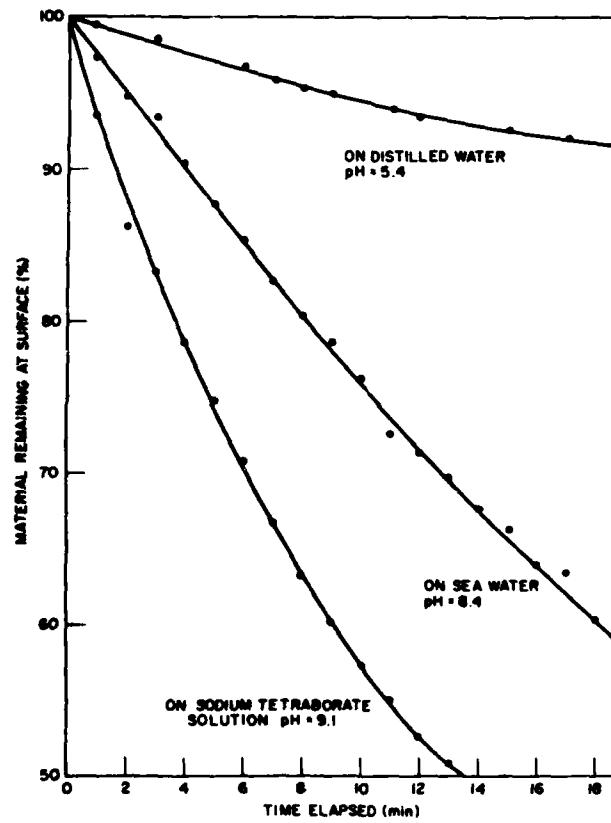


Fig. 4 - Loss of oleic acid from its monolayer
at a constant film pressure of 20.7 dynes/cm

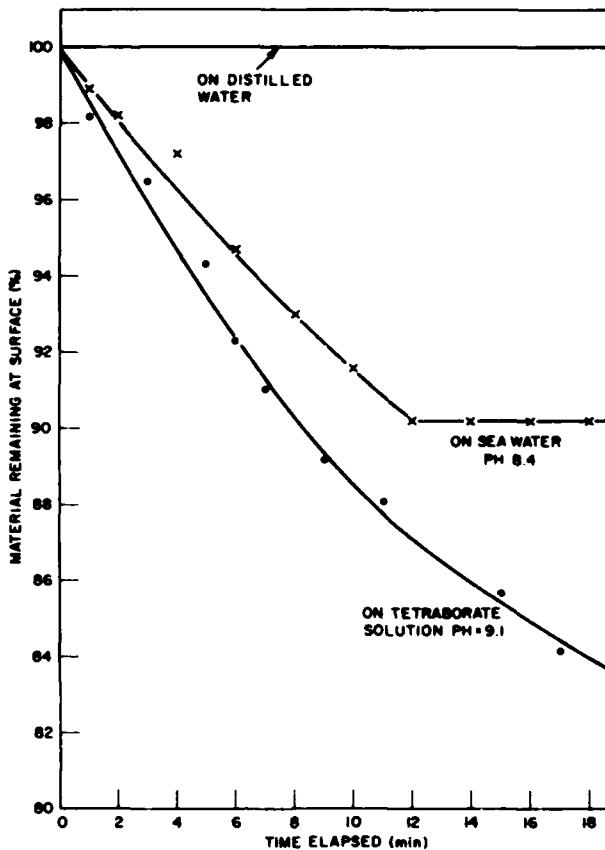


Fig. 5 - Loss of peanut oil from its monolayer at a constant film pressure of 10.3 dynes/cm

CAPILLARY EFFECTS ON WAVELENGTH

The capillary-wave-damping property is not directly related to surface-tension decreases (increases in film pressure), since the wave attenuation rate for a particular wavelength does not continue to rise with increasing film pressure (7). The wave-energy loss is associated with a viscous drag of the underlying hydrogen-bonded water molecules when passing waves force the monolayer-substrate system to compress and dilate. The damping becomes maximum when the water surface has been immobilized by the surface film, and the film pressure begins to increase rapidly with decreasing film area. At this point, about 1-dyne/cm film pressure for water-insoluble monolayers, the film undergoes a transition from a compressible, gaslike state to one of greater coherence and somewhat less compressibility.

However, field observations revealed that surface films with high equilibrium film pressures cause more visible capillary-wave damping than natural sea slicks or artificial films with lower film pressures. An explanation for this effect could not be

attributed to damping of a particular wavelength by the different surface films but was possibly related to wavelength changes which are controlled by surface tension. Vines (17) noted that wavelengths are shortened in the presence of monolayers. He described the progressive increase of capillary-wave damping with decrease in wavelength as the film pressure of a hexadecyl alcohol monolayer was slowly increased. Laboratory studies of the candidate slick materials show the effect of surface tension on capillary wavelength as predicted by an expression derived from the Kelvin equation (18) for the velocity of waves on a liquid in a deep basin:

$$v^2 = \frac{g\lambda}{2\pi} + \frac{2\pi\gamma}{\rho\lambda}, \quad (1)$$

$$v = f\lambda, \quad (2)$$

and

$$\gamma = \frac{\rho f^2 \lambda^3}{2\pi} - \frac{\rho g \lambda^2}{4\pi^2}, \quad (3)$$

where

g = gravitational constant,

v = wave velocity,

λ = wavelength,

γ = surface tension,

ρ = density of water,

and

f = wave frequency.

Goodrich (19) has used Eq. (3) to calculate the surface tension of film-covered surfaces on which waves were generated. Mathematical solutions to Eq. (3) are shown in Fig. 6 along with wavelength data (Table 1) from experiments in which the surface tension was reduced by various monolayers.

For each material studied, 25 μ l of the bulk liquid were added to a clean, distilled-water surface with a micropipet. This quantity was in excess of that required to form a monomolecular layer on the ripple tank. The excess surface-active material remained as a tiny unspread drop of bulk liquid in equilibrium with the monolayer so that the surface tension remained constant throughout the experiment. The surface tensions in Table 2 were measured at 22°C for the different materials by the du Nouy ring method.

Two optical methods were used to determine wavelengths. In the first method capillary waves were generated in a 1-cm-deep glass tray by a triangular Teflon bar attached to a speaker coil. The waves were illuminated from below by light from an electronic stroboscope, and the images of the crests were focused on a screen above the water surface. The same audio oscillator was used to drive both the speaker coil and the stroboscope so that the images of the wave crests appeared as a series of stationary lines of light on the screen. The length of ten waves was measured on the screen while the screen was suspended at heights of 30, 20, and 10 cm above the water surface. The straight line formed by the three points was extrapolated to the surface to give the actual wavelength. Waves were measured by this method at frequencies of 20, 40, 60, 80, 100, and 120 cps.

The second method of wavelength determination was based on the fact that a straight line below the water surface will appear distorted when viewed through the rippled surface at an angle. Capillary waves were generated as in the first method, and the distorted

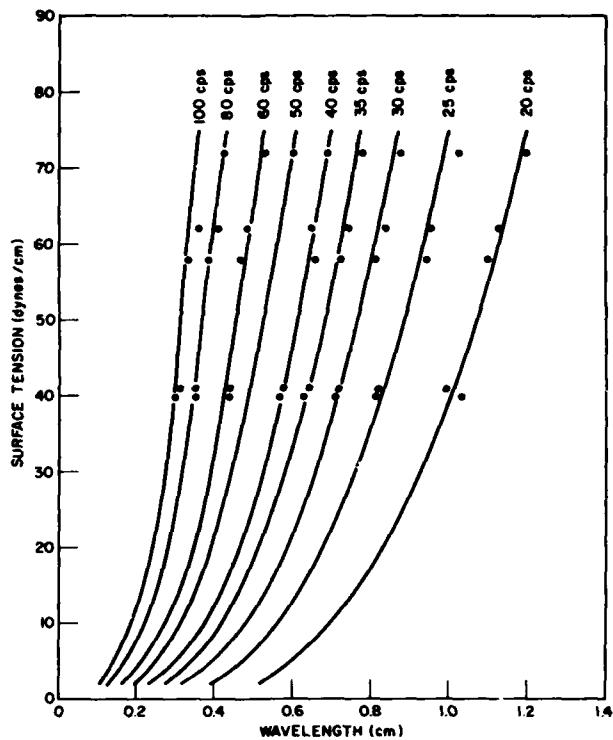


Fig. 6 - Relationship between surface tension and wavelength for capillary waves generated on monolayer-covered surfaces at various frequencies. The curves represent solutions to equation (3).

Table 1
Wavelength Data

F*	Oleyl Alcohol			Oleic Acid			Cottonseed Oil			Tricresyl Phosphate			Water		
	λ^\dagger	V^\ddagger	$1/\lambda$	λ	V	$1/\lambda$	λ	V	$1/\lambda$	λ	V	$1/\lambda$	λ	V	$1/\lambda$
20	1.03	20.6	0.97	0.99	19.8	1.01	1.10	22.0	0.91	1.13	22.6	0.89	1.20	24.0	0.83
25	0.81	20.3	1.23	0.82	20.5	1.22	0.94	23.5	1.06	0.95	23.8	1.05	1.03	25.8	0.97
30	0.71	21.3	1.41	0.72	21.8	1.39	0.81	24.3	1.23	0.84	25.2	1.19	0.88	26.4	1.14
35	0.63	22.1	1.59	0.64	22.4	1.56	0.72	25.2	1.39	0.74	25.9	1.35	0.78	27.3	1.28
40	0.57	22.8	1.75	0.58	23.2	1.72	0.66	26.4	1.52	0.65	26.0	1.54	0.69	27.6	1.45
60	0.44	26.4	2.27	0.44	26.4	2.27	0.47	28.2	2.13	0.49	29.4	2.04	0.53	31.8	1.89
80	0.35	28.0	2.86	0.35	28.0	2.86	0.39	31.2	2.56	0.39	31.2	2.56	0.42	33.6	2.38
100	0.30	30.0	3.33	0.31	31.0	3.23	0.33	33.0	3.03	0.36	36.0	2.78	-	-	-
120	0.27	32.4	3.70	0.28	33.6	3.57	0.29	34.8	3.45	-	-	-	-	-	-

*F = frequency.

† λ = wavelength.

‡V = velocity.

image of the line was photographed along with a scale positioned very close to the surface. Black-and-white 35-mm slides of the wave image and scale were projected onto a screen giving an enlarged image. Measurements were made of waves generated at frequencies of 25, 30, 35, and 40 cps. The combined results of both methods of wavelength determination are presented in Fig. 6.

Table 2
Surface-Tension Measurement by the du Nuoy Ring Method

Monolayer	Surface Tension (dynes/cm)	Equilibrium Film Pressure (dynes/cm)
Clean-water surface	72	0
Tricresyl phosphate	63	9
Cottonseed oil	58	14
Oleic acid	41	31
Oleyl alcohol	40	32

Since shorter waves are more easily damped, the wavelength shift due to surface-tension change is a factor to be considered in the selection of a material for forming an artificial sea slick. A high-pressure monolayer (low surface tension) produces a greater wavelength decrease for a particular sea-surface disturbance. Since shorter waves are attenuated to a greater extent, the high-pressure artificial slick damps a sea surface more intensely and probably resists to a greater degree the formation of small waves.

CONCLUSIONS

Criteria for Monolayer Selection

As consequence of the described laboratory and field experiments, a list of desirable monolayer properties has been compiled which will aid in the selection of a durable and effective sea-surface film. Since evaporation retardation at sea seems unlikely, we are not restricted to the choice of linear molecules which can adlineate at high film pressures. In contrast to this restriction, no special molecular configuration is required for damping capillary waves, and a wide variety of surface-active materials is available. Damping of waves by water-insoluble monolayers becomes maximum at film pressures of only 1 dyne/cm (?) for a particular wavelength. However, substances which spread into monolayers with high film pressures are most desirable because of their wavelength reduction effect which leads to greater wave-damping intensity. The guiding criteria for the selection of a spontaneously spreading monolayer-forming material are given in Table 3.

Table 3
Criteria for the Selection of Artificial Sea-Surface Film

Desirable Property	Chemical Requirement
Nonvolatile, water insoluble	High molecular weight
No chemical interaction with seawater	Un-ionized fatty alcohols
Liquid at most temperatures, rapid spreading for immediate effectiveness	Chemically unsaturated molecule
High film pressure for greatest wave-damping intensity	Film pressures higher than those of glyceryl fatty esters
Commercially available at low cost	Derived from natural products

The fatty acids were judged unsatisfactory, because they are rapidly lost from their monolayer by conversion to soluble soaps by seawater. In addition, they were corrosive to metal and rubber components of the slick-dispersing equipment over a period of time. The natural vegetable oils (mixed-glyceryl, fatty esters) did not create an intensely damped slick, and their effect on air-sea interactions were not as great as that of materials which produced monolayers with higher film pressures. The homologous series of high-molecular-weight, saturated fatty alcohols (solid) possess most of the desired qualities. However, as solids they are not easily dispensed and spread much more slowly than liquids. Consequently, films formed from these materials are not readily re-formed once they have been fragmented by the dispersive action of waves and breaking water.

Of the materials examined, the unsaturated, 18-carbon alcohol, oleyl alcohol (9-octadecene-1-ol, cis isomer), met all of the above requirements. It does not saponify with seawater and has a relatively high equilibrium film pressure (32 dynes/cm at 22°C). The permanently bent molecular structure provided by its cis unsaturated form imparts a liquid character not only to the bulk material but also to its monolayer. As a liquid oleyl alcohol spreads rapidly into a monomolecular layer at temperatures above its freezing point (5°C). Its monolayer behaves like a two-dimensional fluid and adjusts readily to restore monolayer losses caused by surface dilations and other dispersive effects at the air/sea interface. Another compound, sorbitan monooleate, possesses most of the advantages of oleyl alcohol and has a greater film pressure of 40.8 dynes/cm on distilled water at 25°C. The sorbitan monooleate is a fatty acid monoester of sorbitol and has three hydroxyl groups and an oxygen bridge in its structural formula. Thus, it has greater water solubility than oleyl alcohol, giving it a shorter lifetime on the sea surface. The potential usefulness of sorbitan monooleate will be evaluated in future comparative field experiments with oleyl alcohol.

Potential Uses of Artificial Sea Slicks

The role of surface films in modifying micro- and molecular-scale processes at the air/sea interface have been reviewed in the Introduction. It is worthwhile to consider the influence of surface films on geophysical parameters of larger scale, since artificial monolayer-forming substances may someday provide means of producing useful modifications of the marine environment. The primary geophysical properties affected by the microscale effects of ocean films are summarized in Table 4. Several features of the marine environment may be altered by the capillary-wave-damping and wave-resistant characteristics of the immobile films. Reduction of the average-wave slope increases the quantity of light transmitted across the air/water interface and reduces reflected light from the slick-affected areas (Figs. 1 and 2). Slicks should also decrease the form drag of the wind on water, increase wind speed in the boundary layer (20), and reduce breaking waves, although these effects have not been substantiated through definitive experiments at sea. The other listed parameters may have relevance to the fields of cloud physics and oceanography. When these processes are better understood, their modification may be practical in localized situations.

A rationale for the modification of air-sea interactions and exchange processes by artificial monomolecular films lies in the numerous microscale surface properties altered by adsorbed surface-active substances. While it would be presumptuous to undertake massive modifications of the marine environment with these tenuous slicks, many useful effects may be produced in local areas through the application of a coherent, durable surface film.

Table 4
Geophysical Effects of an Artificial Sea Slick

Microscale Modification of Air/Sea Interface	Large-Scale Property Affected
Damping of capillary waves	Light transmission through sea surface; reflection of light and other electromagnetic radiation; wind drag and velocity, wave breaking
Inhibition of convectional cells Bubble-bursting characteristics and sea spray	Surface temperature Production of sea-salt aerosols, transfer of organics from sea to atmosphere
Bubble stability	Sea-foam stability

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14 KEY WORDS	LINK A		LINK B		LINK C	
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Sea-surface films Salt condensation nuclei Air bubbles Air-sea interface Surface-active substances Cloud physics Artificial Sea Slicks Air-Sea Interactions						

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